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PHASE-DISSIPATIVE MECHANISMS FOR LASER-INDUCED SURFACE
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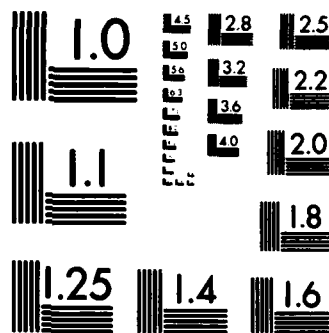
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Task No. NR 056-749

TECHNICAL REPORT No. 56

Phase-Dissipative Mechanisms for Laser-Induced Surface
Desorption/Dissociation Processes

by

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Prepared for Publication

in

The Journal of Physical Chemistry

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October 1984

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1. REPORT NUMBER UROCHESTER/DC/84/TR-56	2. GOVT ACCESSION NO. AD-A149 961	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) Phase-Dissipative Mechanisms for Laser-Induced Surface Desorption/Dissociation Processes		5. TYPE OF REPORT & PERIOD COVERED Interim Technical Report
		6. PERFORMING ORG. REPORT NUMBER
7. AUTHOR(s) Xi-Yi Huang, Thomas F. George, Jian-Min Yuan and L. M. Narducci		8. CONTRACT OR GRANT NUMBER(s) N00014-80-C-0472
9. PERFORMING ORGANIZATION NAME AND ADDRESS Department of Chemistry University of Rochester Rochester, New York 14627		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS NR 056-749
11. CONTROLLING OFFICE NAME AND ADDRESS Office of Naval Research Chemistry Program Code 472 Arlington, Virginia 22217		12. REPORT DATE October 1984
		13. NUMBER OF PAGES 11
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)		15. SECURITY CLASS. (of this report) Unclassified
		15a. DECLASSIFICATION/DOWNGRADING SCHEDULE
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17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)		
18. SUPPLEMENTARY NOTES Prepared for publication in the Journal of Physical Chemistry.		
19. KEY WORDS (Continue on reverse side if necessary and identify by block number) ADMOLECULE-SURFACE SYSTEMS LASER-INDUCED VIBRATIONAL EXCITATION ENERGY-RELAXATION MECHANISMS PHASE-RELAXATION MECHANISMS T ₂ PROCESSES DESORPTION DISSOCIATION QUANTUM MECHANICAL STUDY ZWANZIG PROJECTORS WEIDLICH TECHNIQUE		
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PHASE-DISSIPATIVE MECHANISMS
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Abstract

Vibrational excitation and relaxation for a molecule adsorbed on a surface is investigated in connection with desorption/dissociation processes. The population equations for a laser-driven anharmonic oscillator involving both energy- and phase-relaxation mechanisms are derived with the Zwanzig projector technique and eigenfunction-expansion method due to Weidlich. Phase relaxation is seen to assist in the excitation of the resonant active mode.

I. Introduction

Infrared laser radiation impinging onto a molecule adsorbed on a solid surface can resonantly excite the ad molecular internal vibrational/rotational modes, or deposit photon energy in the ad bond with which the molecule is bound to the surface.¹⁻⁵ It has been shown experimentally^{6,7} that SF_6 and CH_3F molecules adsorbed on NaCl surfaces at low temperatures can be desorbed by the resonant excitation of the adsorbate internal vibration, namely, the ν_3 mode. The treatment of the problem of many degrees of freedom for the molecule-surface system is very complicated and generally an insoluble problem. Interactions between the resonant active vibrational mode and the other inactive modes, and between the ad molecule and surface excitations - surface phonons, surface plasmons, reflected photons (i.e., photons emitted by the ad molecule and reflected by the surface), etc. - create a large number of dissipative channels by which absorption, dissociation, desorption or chemical reaction on the surface are strongly influenced. (Desorption is the breaking of the ad bond between the ad molecule and the surface, and dissociation is the breaking of a bond within the ad molecule.) In this Letter, we are especially interested in the influence of surface-dephasing relaxation on the excitation of the resonant mode, which plays an important role in the process of desorption/dissociation.

II. Theory

Under normal circumstances, due to the vast number of degrees of freedom of the ad molecule-surface system, the intrasystem relaxation can be very fast, and in fact, comparable to or faster than the rate of photon absorption. In this case, energy randomization or statistical thermodynamical behavior may at least partly occur in desorption/dissociation processes on surfaces.⁸ The IR-laser-driven

resonant active mode absorbs photons and transfers its energy to other vibrational modes or the surface, and then "interference" superposition of these many vibrations can instantaneously accumulate energy in the weakest bond to lead to molecule dissociation or desorption from the surface, if this energy is beyond the appropriate threshold.^{9,10} As an example, the threshold temperature for thermal dissociation of SF_6 in the gas phase is measured to be 1600 K.⁸ At this temperature, the average occupation number of the resonant mode (ν_3) of SF_6 is $\bar{n}=0.76$, which means that, while the energy deposited in the molecule has gone into the ν_3 mode to exceed the critical value for dissociation, the other modes remain in low excited states.

We shall confine ourselves here to the excitation and relaxation of the active anharmonic vibrational mode which is driven by a resonant coherent field and controlled by both energy (T_1) and phase (T_2) relaxation. For this purpose, we treat the many other non-active modes and surface excitations (i.e., phonons, plasmons, reflected photons, etc.) as a large thermal reservoir, by which dissipation mechanisms are provided. A reduced density operator W for the resonant mode obeys the master equation

$$\begin{aligned} \frac{dW}{dt} &= \left[\frac{dW}{dt} \right]_{\text{coherent}} + \left[\frac{dW}{dt} \right]_{\text{incoherent}} \\ &= \left[-iL_c + \Lambda_1 + \Lambda_2 \right] W. \end{aligned} \quad (1)$$

The reversible term is $-iL_c W \equiv -(i/\hbar)[H, W]$, where $H = H_M + H_F$ is the sum of anharmonic oscillator and external field interactions,¹¹

$$H_M = \hbar(\omega - \omega_L) a^\dagger a - \hbar \epsilon a^\dagger a (a^\dagger a + 1) \quad (2)$$

$$H_F = \mu E(t)(a^\dagger + a). \quad (3)$$

Here a^\dagger and a are oscillator ladder operators, μ is the molecular dipole moment, $E(t)$ is the applied electric field, ω_L is the laser frequency, and ϵ is a positive anharmonicity. The dissipative terms are given by^{12,13}

$$\begin{aligned} \Lambda_1 W = & \gamma_\downarrow([a, Wa^\dagger] + [aW, a^\dagger]) \\ & + \gamma_\uparrow([a^\dagger, Wa] + [a^\dagger W, a]), \end{aligned} \quad (4)$$

$$\Lambda_2 W = \eta([a^\dagger a, Wa^\dagger a] + [a^\dagger aW, a^\dagger a]). \quad (5)$$

$\Lambda_2 W$ is responsible for a pure dephasing in the decay of the off-diagonal matrix elements,¹⁴ and γ_\uparrow , γ_\downarrow and η contain implicit information about the reservoir, including the surface. With the dissipative terms [Eqs. (4) and (5) only], the relaxation behavior of the expectation values of the amplitude and number operator is governed by the equations $\frac{d}{dt}\langle a \rangle = -(\gamma_\downarrow - \gamma_\uparrow + \eta)\langle a \rangle$ and $\frac{d}{dt}\langle a^\dagger a \rangle = 2(\gamma_\downarrow - \gamma_\uparrow)\langle a^\dagger a \rangle + 2\gamma_\uparrow$. Λ_2 thus affects only the off-diagonal elements of the density operator by providing a pure phase-destroying term with a rate of η . In the Born approximation, the diagonal parts of the density matrix obey the equation

$$\frac{d}{dt} W_n = \langle n | \Lambda_1 W_r | n \rangle - \int_0^t d\tau \langle n | P L_i e^{-iL_M(t-\tau)} e^{\Lambda_2(t-\tau)} L_i W_r(\tau) | n \rangle, \quad (6)$$

where L_i is a Liouville operator due to the oscillator-field interaction, P is a projection operator used to obtain the diagonal part of the density operator,¹⁵ and $W_r \equiv PW$ is the relevant part of the density matrix.

The difficult problem here is the explicit evaluation of the matrix elements that appear inside the integral in Eq. (6). We use the Weidlich eigenfunction-expansion method^{12,13} to evaluate these elements. The "eigenstate" of the super-operators Λ_2 and Λ_2^\dagger are $||A_{pq}\rangle$ and $||B_{pq}\rangle$, respectively, and under the Markoff assumption, the "eigenvalues" of Λ_2 have the form $\lambda_{pq} \approx -\eta q^2$, where $p = 0, 1, 2, \dots$ and $q = 0, \pm 1, \pm 2, \dots$. The equations of motion for the diagonal parts of the density matrix are then given as

$$\dot{W}_n = \langle n | \dot{A}_1 W_r | n \rangle - \int_0^t d\tau \sum_{pq} e^{-nq^2(t-\tau)} \times \text{Tr}(B_{pq}^+ L_i W_r(\tau)) \langle n | L_i e^{-iL_M(t-\tau)} A_{pq} | n \rangle. \quad (7)$$

By a lengthy but straightforward algebraic calculation, we obtain a simpler set of coupled differential equations to describe the excitation and relaxation behavior for the resonant vibrational mode:

$$\dot{W}_n = 2\kappa [(n+1) W_{n+1} - n W_n] - 2\eta \left[\frac{\Omega_R(t)}{\eta} \right]^2 \left\{ \frac{n(W_n - W_{n-1})}{1 + [2\frac{\epsilon}{\eta}(n-1) - \frac{\Delta}{\eta}]^2} + \frac{(n+1)(W_n - W_{n+1})}{1 + (2\frac{\epsilon}{\eta}n - \frac{\Delta}{\eta})^2} \right\}. \quad (8)$$

The decay parameters κ ($\equiv \gamma_{\downarrow} - \gamma_{\uparrow}$) and η are determined by the internal mode-mode coupling and the ad molecule-surface interaction, and for a fixed temperature and ad molecule-surface distance they can be considered as constants (here a cold surface of 0 K has been assumed), $\Omega_R(t)$ is the Rabi frequency, and Δ is the detuning between the ad molecular fundamental and laser frequencies.

The first two terms with the factor of 2κ on the right-hand side of Eq. (8) describe the energy relaxation processes which always lead to population loss in the resonant mode. In the next terms with the factors of η describing the influence of the phase relaxation, we notice that there are competitive factors: upon an increase of the value of η , the phase relaxation tends to diminish the detuning and anharmonicity "bottleneck" effects, and the effective Rabi frequency $\Omega_R/\eta^{1/2}$ tends to decrease.

III. Calculations and Results

Figure 1 shows the influences of the phase relaxation. The time evolution for the average occupation number \bar{n} of a driven damped anharmonic oscillator is displayed, with consecutive values of the phase-relaxation parameter η ($\eta = 1, 5, 9, 13$). We note that there is a range of values of η which assist the excitation of the ad molecular active mode. For the optimal parameter $\eta = 5$ in our specific case, Figure 2 displays the time-dependent population distributions for several excited states, where considerable population is seen. One would generally expect the phase decay η to be much more enhanced than κ by the inactive adspecies vibrational modes and by the surface.¹⁶ The phase-dissipation mechanism may be generated in several ways in an ad molecule-surface system as follows: (a) intramolecular coupling between the active and inactive vibrational modes;¹⁷ (b) intermolecular collisions when the ad molecules can migrate on the surface;¹⁸ (c) rotational relaxation within the vibrational level manifold of the ad molecule;¹¹ (d) self-coupling of the ad molecule due to surface-reflected photons.^{19,20}

Acknowledgments

This research was supported in part by the Air Force Office of Scientific Research (AFSC), United States Air Force, under Grant AFOSR-82-0046, and the Office of Naval Research. The United States Government is authorized to reproduce and distribute reprints for governmental purposes notwithstanding any copyright notation hereon. TFG acknowledges the Camille and Henry Dreyfus Foundation for a Teacher-Scholar Award (1975-84).

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Figure Captions

1. Time dependence of the average occupation number $\bar{n} = \langle a^\dagger a \rangle$ for (cw) laser-driven anharmonic oscillator in contact with a bath. The curves correspond to $\eta = 1, 5, 9, 13$, and the others parameters are $(\kappa, \Omega_R, \epsilon, \Delta) = (0.01, 0.5, 1.5, 3.0)$, where all rates are in the unit of the transverse relaxation¹³ rate. This figure shows that when the vibrational linewidth η is smaller than the detuning Δ , the maximum vibrational excitation is small but grows as η increases. Excessively large values of η eventually reduce the average excitation level, presumably due to the spread of population within the linewidth.
2. The probability of occupation of level n , $W_n(t)$, as a function of time. η is fixed at 5, and the other parameters are given in Figure 1.

Figure 1

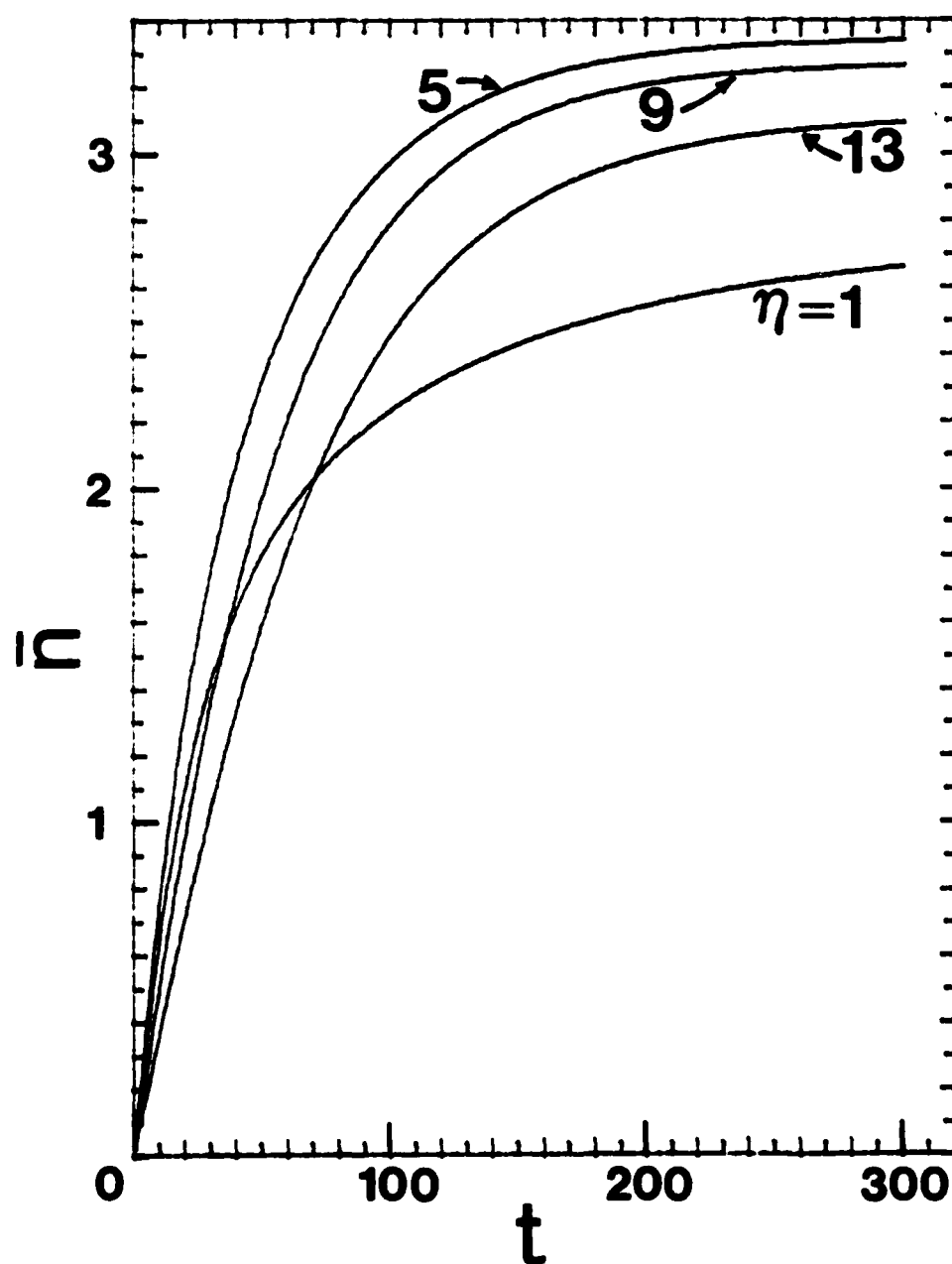
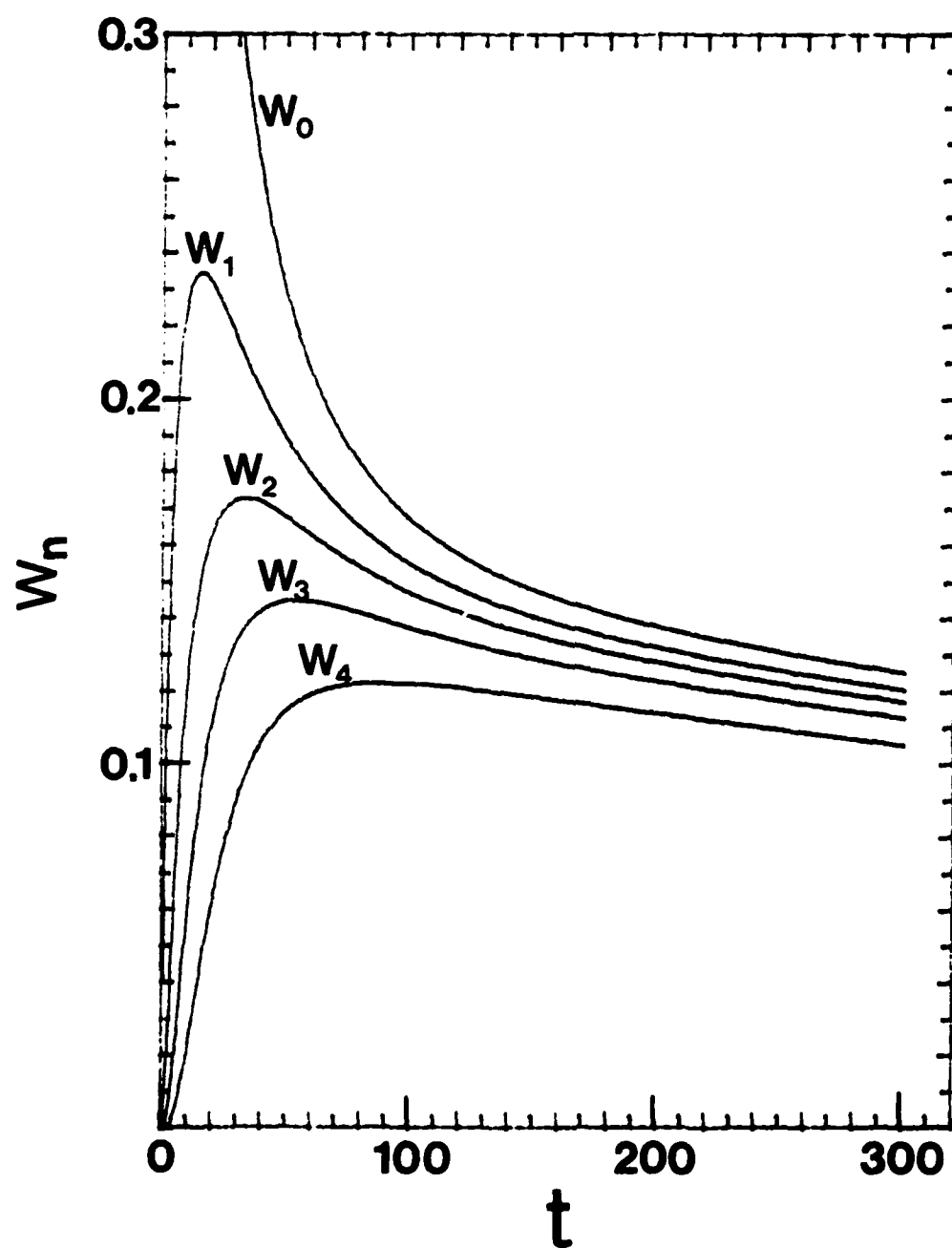


Figure 2



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